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| FORM PTO-1390 (Modified)<br>(REV 10-95)  |   | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE     | ATTORNEY'S DOCKET NUMBER<br><b>AD-6705</b>                      |
| <b>TRANSMITTAL LETTER TO THE UNITED STATES<br/>DESIGNATED/ELECTED OFFICE (DO/EO/US)<br/>CONCERNING A FILING UNDER 35 U.S.C. 371</b>  |   |   | U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR)<br><b>09/890028</b> |
| INTERNATIONAL APPLICATION NO.<br><b>PCT/US00/04754</b>   | INTERNATIONAL FILING DATE<br><b>25 FEBRUARY 2000 (25.02.00)</b> | PRIORITY DATE CLAIMED<br><b>25 FEBRUARY 1999 (25.02.99)</b> |   |
| TITLE OF INVENTION<br><b>AQUEOUS DISPERSION COMPOSITION AND MANUFACTURING METHOD FOR THE COMPOSITION</b>   |   |   |   |
| APPLICANT(S) FOR DO/EO/US<br><b>NAKATA, Kazuyuki</b>   |   |   |   |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information   |   |   |   |
| <ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input checked="" type="checkbox"/> This is an express request to being national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b)) and PCT Articles 22 and 39(1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19<sup>th</sup> month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application was filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau.</li> <li>b. <input type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</li> </ol> </li> <li>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371 (c) (2)).</li> <li>7. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210).</li> <li>8. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c) (3)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input checked="" type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>9. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).</li> <li>10. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).</li> <li>11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409)</li> <li>12. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).</li> </ol> |   |   |   |
| Items 13 to 18 below concern document(s) or information included :   |   |   |   |
| <ol style="list-style-type: none"> <li>13. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>15. <input type="checkbox"/> A <b>FIRST</b> preliminary amendment.<br/>A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li>16. <input type="checkbox"/> A substitute specification.</li> <li>17. <input checked="" type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>18. <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail.</li> <li>19. <input type="checkbox"/> Other items or information:</li> </ol>  |   |   |   |
| <div style="border: 1px solid black; padding: 5px;"> <b>17. General Power of Attorney</b><br/> <b>18. Express Mailing Label No.: EL031053745US</b> </div>  |   |   |   |

|   |                     |  |  |  |    |
|---|---------------------|--|--|--|----|
| APPLICATION NO. (IF KNOWN, SEE 37 CFR)<br><b>09/890028</b>  |                     | INTERNATIONAL APPLICATION NO.<br><b>PCT/US00/04754</b> |  | ATTORNEY'S DOCKET NUMBER<br><b>AD-6705</b> |    |
| 20. The following fees are submitted  |                     |  |  | <b>CALCULATIONS PTO USE ONLY</b>           |    |
| <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :</b>  |                     |  |  |  |    |
| <input checked="" type="checkbox"/> Search Report has been prepared by the EPO or JPO   |                     | \$860.00   |  |  |    |
| <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482)   |                     | \$690.00   |  |  |    |
| <input type="checkbox"/> No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2))  |                     | \$760.00   |  |  |    |
| <input type="checkbox"/> Neither international preliminary examination fee paid to USPTO (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO   |                     | \$1000.00  |  |  |    |
| <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) And all claims satisfied provisions of PCT Article 33(2)-(4)  |                     | \$ 100.00  |  |  |    |
| <b>ENTER APPROPRIATE BASIC FEE AMOUNT</b> =   |                     |  |  | <b>\$860.00</b>                            |    |
| Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30   |                     |  |  | <b>\$0.00</b>                              |    |
| <b>CLAIMS</b>   | <b>NUMBER FILED</b> | <b>NUMBER EXTRA</b>                                    | <b>RATE</b>  |  |    |
| Total Claims  | <b>18 - 20 =</b>    | <b>0</b> x   | \$18.00  | <b>\$0.00</b>                              |    |
| Independent Claims  | <b>3 - 3 =</b>      | <b>0</b> x   | \$80.00  | <b>\$0.00</b>                              |    |
| Multiple Dependent Claims (check if applicable)   |                     |  | <input checked="" type="checkbox"/>  | <b>\$270.00</b>                            |    |
| <b>TOTAL OF ABOVE CALCULATIONS</b> =  |                     |  |  | <b>\$270.00</b>                            |    |
| Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable).   |                     |  | <input type="checkbox"/>   | <b>\$0.00</b>                              |    |
| <b>SUBTOTAL</b> =   |                     |  |  | <b>\$270.00</b>                            |    |
| Processing Fee of <b>\$130.00</b> for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30  |                     |  |  | <b>\$0.00</b>                              |    |
| <b>TOTAL NATIONAL FEE</b> =   |                     |  |  | <b>\$1,130.00</b>                          |    |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).   |                     |  | <input type="checkbox"/>   | <b>\$0.00</b>                              |    |
| <b>TOTAL FEES ENCLOSED</b> =  |                     |  |  | <b>\$1,130.00</b>                          |    |
|   |                     |  |  | Amount to be : refunded                    | \$ |
|   |                     |  |  | Charged                                    | \$ |
| <input type="checkbox"/> A check in the amount of _____ to cover the above fees enclosed.<br><input checked="" type="checkbox"/> Please charge my Deposit Account No. <b>04-1928</b> in the amount of <b>\$1,130.00</b> to cover the above fees.<br><input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. <b>04-1928</b> a duplicate copy of this sheet is enclosed. |                     |  |  |  |    |
| <b>NOTE : Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (CFR 1.37(a) or (b)) must be filed and granted to restore the application to pending status.</b><br><b>SEND ALL CORRESPONDENCE TO:</b>   |                     |  |  |  |    |
| <b>EVANS, Craig H.</b><br><b>E. I. DU PONT DE NEMOURS AND COMPANY</b><br><b>Legal Patent Records Center</b><br><b>1007 Market Street</b><br><b>Wilmington, Delaware 19898</b><br><b>United States of America</b>  |                     |  | <div style="text-align: center;"> <br/>           SIGNATURE<br/> <b>EVANS, CRAIG H.</b><br/>           NAME<br/> <b>31,825</b><br/>           REGISTRATION NUMBER<br/> <b>July 20, 2001</b><br/>           DATE         </div> |  |    |

5

TITLEAQUEOUS DISPERSION COMPOSITION AND MANUFACTURING  
METHOD FOR THE COMPOSITIONFIELD OF THE INVENTION

The present invention relates to an aqueous dispersion  
composition having good stability and dispersion properties and a process for  
making it by dispersing an ethylene-methacrylic acid copolymer in water using an  
amount of ammonia in excess of amount that would be needed to neutralize  
methacrylic acid. It also relates to the application of this aqueous dispersion in  
making coated substrates.

BACKGROUND OF THE INVENTION

Aqueous dispersion compositions of ethylene  $\alpha,\beta$ -ethylenically  
unsaturated carboxylic acid copolymers such as ethylene-acrylic acid copolymer  
or ethylene-methacrylic acid copolymer are known and sold commercially. They  
can be easily made by dispersing the readily available ethylene acid copolymers  
in water using an alkali metal compound and are useful in various applications  
such as coating film. However, because the film coated with such dispersion  
compositions has a poor waterproofness, it cannot be used for applications  
requiring a film that is waterproof.

It is known that an ethylene-acrylic acid copolymer can be used as  
an excellent raw material for an aqueous dispersion composition obtained using  
ammonia as dispersion aide, see for example U.S. 3,674,896 and GB 2,269,822.  
Although it has been difficult to obtain an aqueous dispersion composition using  
ammonia alone, a film coated with such aqueous dispersion should have a good  
waterproofness since there is no alkali ion to absorb water.

GB 1,559,048 describes an aqueous dispersion of ethylene-  
methacrylic acid copolymer partially neutralized with sodium ions and optionally  
residual 90% with ammonium ions for coating substrates.

Uniformly dispersing ethylene-methacrylic acid copolymer in  
water using a mixture of ammonia and an alkali metal has been known to be  
difficult. Such aqueous dispersions of ethylene-methacrylic acid copolymer  
could be obtained by adding small amounts of a surfactant as a supplemental  
disperser. However, because the coated film using such dispersions have both  
poor waterproofness and bleed-out, causing contamination, it was not preferred.

An object of this invention is to find a way to use ammonia alone  
as a dispersion aide to make a uniform aqueous dispersion composition of an  
ethylene-methacrylic acid copolymer that is stable for a long time, that is has a

- 5 good shelf-life of say a year or more. Another object was to find a way to make a film (or other substrate) that when coated with such an aqueous dispersion would have good waterproofness.

## 5 SUMMARY OF THE INVENTION

Attempts to obtain such an aqueous dispersion of ethylene-methacrylic acid copolymer using ammonia in an amount equivalent to that of the carboxyl groups contained in an ethylene-methacrylic acid copolymer were unsuccessful in making uniform dispersions.

10 With continued research, it was found that a stable, uniform aqueous dispersion of ethylene-methacrylic acid copolymer could be obtained using an ethylene-methacrylic acid copolymer containing specific amounts of acid, and using ammonia in an amount greater than the amount of the carboxyl groups in the copolymer. It was also found that coating a film (or other substrate)  
15 with such an aqueous dispersion could make a waterproof, coated film (or other substrate).

The present invention, claiming priority to Japanese Patent Application No. Hei 11[1999]-48872 which is incorporated herein by reference, relates to a process for making a uniform aqueous dispersion of ethylene-methacrylic acid copolymer having good dispersion stability and to the aqueous  
20 dispersion made from such process. It also relates to a coated substrate such as a film that, when coated with the aqueous dispersion of the present invention, has good waterproof properties. It also relates to a laminate obtained by applying the above-mentioned aqueous dispersion composition on a substrate for coating and  
25 drying to form a coated substrate.

The stable, uniform aqueous dispersion of the present invention consists essentially of a dispersion of component (A), an ethylene-methacrylic acid copolymer containing 15-35 wt% of methacrylic acid, and component (B), ammonia in an amount required for neutralizing 110-150% of the carboxyl  
30 groups of component (A) in water. The ethylene-methacrylic acid copolymer preferably comprises 5-50 wt% of the dispersion and preferably has a melt flow rate of 50-2000 grams/10 minutes at 190°C/2160 gram load. Surfactants and the like are not needed and preferably are not used.

## DETAILED DESCRIPTION OF THE INVENTION

35 "Consisting essentially of" means that the recited components are essential, while smaller amounts of other components may be present to the extent that they do not detract from the operability of the present invention.

"Copolymer" means polymers containing two or more monomers.

A stable, uniform ethylene acid-containing copolymer aqueous  
40 dispersion composition can be obtained by mixing selected ethylene methacrylic acid copolymer in water in the presence of an excess amount of ammonia.

5           The ethylene acid-containing copolymer aqueous dispersion of the present invention consists essentially of a dispersion of component (A), an ethylene-methacrylic acid copolymer containing 15-35 wt% of methacrylic acid as the ethylene acid-containing copolymer, in water in the presence of component (B), ammonia used as a basic component in an amount greater than the amount of  
10   the carboxyl groups of component (A).

Both good dispersion property and good dispersion stability can be obtained by using an excess of component (B) ammonia, particularly an amount sufficient for neutralizing 110-150% of the carboxyl groups of the above-mentioned acid-containing copolymer (A). The resulting aqueous dispersion can  
15   be coated onto a substrate, such as a film, to make a coated substrate, particularly a coated film, that is not susceptible to moisture accumulation and has a good waterproofness.

It is suitable for ethylene-methacrylic acid copolymer (A) to contain 15-35 wt% or alternatively 15-25 wt%, particularly 18-30 wt%, of an  
20   unsaturated carboxylic acid. In the case of using a copolymer containing an unsaturated carboxylic acid in an amount that is less than the above-mentioned range, it is difficult to obtain a composition having a good aqueous dispersion property. In the case of using a copolymer containing an unsaturated carboxylic acid in an amount that is more than the above-mentioned range, a stable  
25   dispersion composition cannot be obtained and both the waterproofness and mechanical strength of the coated film are reduced.

An ethylene-methacrylic acid copolymer having a melt flow rate of 50-2000 grams/10 minutes, particularly 60-1500, at 190°C/2160 gram load is suitable. In the case of using a methacrylic acid copolymer having an extremely  
30   low melt flow rate, an aqueous dispersion composition having a good dispersion property cannot be obtained. When using a copolymer having an excessively high melt flow rate, the coated film has a poor strength.

Besides ethylene and methacrylic acid, the copolymer may be copolymerized with other monomers including an unsaturated carboxylic acid  
35   ester such as methyl acrylate, ethyl acrylate, n-butyl acrylate, isobutyl acrylate, methyl methacrylate, isobutyl methacrylate, dimethyl maleate, or diethyl maleate; a vinyl ester such as vinyl acetate or vinyl propionate; and carbon monoxide, in an amount of 20 wt% or less, particularly 10 wt% or less.

The aqueous dispersion composition of the present invention  
40   contains ammonia, which can neutralize an excess, particularly 110-150%, more particularly 120-140% of the carboxyl groups of copolymer (A), along with copolymer (A).

5 In the case of an aqueous dispersion composition of the present invention containing a lower amount of ammonia than the above-mentioned range, a stable aqueous dispersion composition cannot be obtained. In the case of an aqueous dispersion composition of the present invention containing a higher amount of ammonia than the above-mentioned range, gelling easily occurs.

10 The aqueous dispersion composition suitably has the copolymer (A) present in an amount of 5-50 wt%, preferably 5-30 wt%, and particularly 10-30 wt%.

The aqueous dispersion composition is obtained by introducing both ethylene-methacrylic acid copolymer (A) and ammonia (B) with water into a  
15 vessel, then stirring them at about 90 to about 150°C for a sufficient time to uniformly disperse the ethylene-methacrylic acid copolymer (A), preferably about 10 minutes to about 2 hours.

The aqueous dispersion composition has good stability and good shelf life, such that neither the particle size nor the viscosity is significantly  
20 changed over times of up to a year or more.

### Additives

A variety of additives can be added to the aqueous dispersion composition if desired.

Examples of additives include polyalcohols such as glycerin,  
25 ethylene glycol, polyethylene glycol, and polypropylene glycol; lower alcohols such as water-soluble epoxy compounds, methanol, ethanol, n-propanol, or isopropanol; ethers such as ethylene glycol monomethyl ether, ethylene glycol monoethyl ether, propylene glycol monomethyl ether, propylene glycol diethyl ether, diethylene glycol monoethyl ether, and dipropylene glycol monomethyl  
30 ether; esters such as propylene glycol monoacetate and ethylene glycol monoacetate; antioxidants; weather resistant stabilizers; ultraviolet-ray absorbents; antistatic agents; pigments; dyestuffs; antibacterial agents; lubricants; inorganic fillers; blocking preventing agents; and adhesives.

### Other Polymer Aqueous Dispersion Compositions

35 The aqueous dispersion composition of the present invention may be mixed with other polymer aqueous dispersion compositions ("Other Dispersions") with a weight ratio based on the solid components in the dispersions of about 10/90 to about 90/10, particularly 20/80-80/20.

The Other Dispersion(s) with which the aqueous dispersion  
40 composition of the present invention may effectively be mixed should have a pH of 7 or more. If the pH of the Other Dispersion(s) is less than 7, it should be

5 adjusted with ammonia to obtain a pH of 7 or more before mixing. Also, the  
Other Dispersion should be one that is not gelled when it is mixed with the  
aqueous dispersion composition of the present invention. It is suitable to choose  
Other Dispersion(s) having an average particle size of 1-10000, preferably 1-  
1000, particularly 5-500 nanometers (nm), and having a solid component amount  
10 of 2-60 wt%, particularly 5-50 wt% of the dispersion. Preferably the particle size  
of the Other Dispersion(s) should be substantially the same as the dispersion of  
the present invention.

Examples of the Other Dispersion(s) include aqueous dispersions  
of ethylene-acrylic acid copolymer (particularly those made using ammonia alone  
15 as dispersing agent), polyvinyl acetate, ethylene-vinyl acetate copolymer,  
polyvinyl chloride, polyvinylidene chloride, water-soluble acryl resins,  
acrylamide resins, methacrylamide resins, acrylonitrile resins, styrene-acrylic  
acid copolymer, water-soluble polyurethane resins, water-soluble styrene-maleic  
acid copolymers, water-soluble polyurethane resins, styrene-butadiene  
20 copolymers, high-impact polystyrene resins, butadiene resins, polyester resins,  
acrylonitrile-butadiene copolymers, polyethylene resins, polyethylene oxide  
resins, polypropylene-ethylene copolymers, maleic anhydride graft-  
polypropylene-ethylene copolymers, polyethylene chloride, polypropylene  
chloride, EPDM (ethylene-propylene-diene polymer), polypropylene chloride,  
25 phenol resins, silicone resins, and epoxy resins. One or more types of these may  
be used.

A mixed aqueous dispersion composition can be obtained by  
stirring and mixing the two or more dispersion compositions at standard  
temperature. The base resin of the dispersion composition of the present  
30 invention may be melt-blended or dry-blended beforehand with the base resin of  
the other dispersion composition to be mixed with it, followed by dispersion in  
water. The present invention is not specifically restricted by any manufacturing  
method.

### Coatings on Substrates

35 The aqueous dispersion composition of the present invention or  
mixed aqueous dispersion composition (aqueous dispersion composition of the  
present invention with Other Dispersion(s)) can be applied on any type of  
substrate as a coating. The coated substrate, particularly a coated film, made by  
this method has a good waterproofness.

40 The aqueous dispersions can be applied to a substrate for coating  
using a conventional method such as that using a roll coater or a bar coater, a  
method involving spraying, a method using an air-knife coater, a method using a

- 5 brush, or a method involving a substrate in the aqueous dispersion. Water is evaporated and a uniform film can be obtained by heating and drying after coating.

Examples of the substrate include molded products made by molding an olefin copolymer such as high, medium, or low-density polyethylene, ethylene- $\alpha$ -olefin copolymer, ethylene-vinyl acetate copolymer, ethylene-(meth)acrylic ester copolymer, ethylene-(meth)acrylic acid copolymer or ionomer, ethylene-(meth)acrylic acid-(meth)acrylic ester copolymer or ionomer, polypropylene, poly-1-butene, or poly-4-methyl-1-pentene; styrene resins such as polystyrene, ABS resin, or styrene-butadiene block copolymer; polyesters such as polyethylene terephthalate; polyamides such as nylon 6 or nylon 66; polyvinyl chloride; or their blends; natural materials such as a film, metal (iron, copper, aluminum, or stainless steel), wood, or paper; natural or synthetic leather; fibers such as nylon, polyester, acryl, urethane, or rayon; and fabrics.

The thickness of the coated film is not specifically restricted, but is suitably 1-20 micrometers ( $\mu\text{m}$ ), particularly 1-5  $\mu\text{m}$ . The coated film may be crosslinked by irradiating using an electron beam to improve the waterproofness or durability.

## EXAMPLES

The following examples are illustrative of the present invention.

25 All parts in the following examples are based on weight.

### 1. Raw materials and additives

The ethylene-methacrylic acid copolymers ("Copolymers") used in the examples are described in Table I. The ammonia used in the experiments was a 29% aqueous solution made by Kanto Kagaku K.K.

30

Table I

| Composition  |   | FR<br>(dg/min) |
|--------------|---|----------------|
| Copolymer 1: | Ethylene/methacrylic acid (80 wt%/20 wt%) | 60             |
| Copolymer 2: | Ethylene/methacrylic acid (80 wt%/20 wt%) | 300            |
| Copolymer 3: | Ethylene/methacrylic acid (80 wt%/20 wt%) | 500            |

### 2. Methods for evaluating the properties

Properties of the dispersions set forth in Table III were determined as follows:

- 5     1. Appearance:     The measurement of Appearance of the resulting  
dispersions was by visual examination. The  
Appearance was considered to be
- a. "Uniform"     if the dispersion contained no non-dispersed substances  
and remained stable for an extended period of time;
- 10     b. "Non-uniform" if the dispersion contained non-dispersed substances or  
precipitates; and
- c. "Not dispersed" if the substances did not disperse or remain dispersed  
in the aqueous media.
- 15     2. pH:     Based on JIS K6833. Obtained by measurement of a  
sample using a commercial pH meter Horiba Custany  
LAB "F12" pH meter.
3. Viscosity:     Based on JIS K6833. Obtained by measurement using  
a single-cylinder rotary viscometer Synchro Lectric  
Viscometer.
- 20     4. Average Particle size:     Measured by laser beam scattering-type particle  
size distribution measuring apparatus NICOMP  
370HPL.

### APPLICATION EXAMPLES 1-3

- 25     Copolymers, ammonia, and distilled water were introduced into an  
autoclave (300 mL) in the amounts shown in Table II, followed by stirring at  
150°C for 60 minutes. The weight percent solid component in the aqueous  
mixture in each case was 25 wt%. Evaluation results for dispersion properties are  
shown in Table III.

### COMPARATIVE EXAMPLES 1-4

- 30     Ammonia was used in the amounts shown in Table II in the same  
process used in Application Examples 1-3. Evaluation results for the dispersion  
property are shown in Table III.

5

Table II

| Composition           |             |  |
|-----------------------|-------------|--|
|                       | Resin       | Neutralization degree (NH <sub>3</sub> ) (%) |
| Application Example 1 | Copolymer 1 | 130  |
| Application Example 2 | Copolymer 2 | 130  |
| Application Example 3 | Copolymer 3 | 130  |
| Comparative Example 1 | Copolymer 1 | 100  |
| Comparative Example 2 | Copolymer 2 | 100  |
| Comparative Example 3 | Copolymer 3 | 100  |
| Comparative Example 4 | Copolymer 1 | 75   |
| Comparative Example 5 | Copolymer 3 | 75   |

Table III

|                       | Appearance    | pH   | Viscosity | Average particle size |
|-----------------------|---------------|------|-----------|-----------------------|
| Application Example 1 | Uniform       | 11.1 | 225       | 53                    |
| Application Example 2 | Uniform       | 11.1 | 400       | 22                    |
| Application Example 3 | Uniform       | 11.0 | 705       | 24                    |
| Comparative Example 1 | Not dispersed |      |           |                       |
| Comparative Example 2 | Non-uniform   | 10.9 | 370       | 26                    |
| Comparative Example 3 | Non-uniform   | 11.0 | 555       | 29                    |
| Comparative Example 4 | Not dispersed |      |           |                       |
| Comparative Example 5 | Not dispersed |      |           |                       |

5 **WHAT IS CLAIMED IS:**

1. A stable, uniform, and alkali metal free aqueous dispersion consisting essentially of a dispersion in water of component (A), an ethylene-methacrylic acid copolymer containing 15-35 wt% methacrylic acid, and component (B), ammonia in an amount required for neutralizing 110-150% of the carboxyl groups of component (A).  
10
2. The aqueous dispersion of claim 1 wherein the ethylene-methacrylic acid contains 18-30 wt% methacrylic acid.
3. The aqueous dispersion of claim 1 wherein the ethylene-methacrylic acid contains 15-25 wt% methacrylic acid.  
15
4. The aqueous dispersion of claims 1, 2, or 3 wherein the ammonia is present in an amount sufficient to neutralize 120-140% of the carboxyl groups.  
20
5. The aqueous dispersion of claim 1 wherein the ethylene-methacrylic acid copolymer comprises 5-50 wt% of the dispersion and preferably has a melt flow rate of 50-2000 grams/10 minutes at 190°C/2160 gram load.
6. The aqueous dispersion of claim 3 wherein the ethylene-methacrylic acid copolymer comprises 5-50 wt% of the dispersion and preferably has a melt flow rate of 50-2000 grams/10 minutes at 190°C/2160 gram load.  
25
7. The aqueous dispersion of claim 4 wherein the ethylene-methacrylic acid copolymer has a melt flow rate of 60-1500 grams/10 minutes at 190°C/2160 gram load.  
30
8. The aqueous dispersion of claim 5 wherein the ethylene-methacrylic acid copolymer has a melt flow rate of 60-1500 grams/10 minutes at 190°C/2160 gram load.  
35
9. A coated substrate obtained by applying the aqueous dispersion of claim 1, 2 or 3 to the substrate for coating, then drying to form a coated substrate.
10. The coated substrate of claim 9 wherein the substrate is a film.  
40

- 5 11. A process for making a stable, uniform, and alkali metal free aqueous dispersion of ethylene-methacrylic acid consisting essentially of mixing an ethylene-methacrylic acid copolymer containing 15-35 wt% methacrylic acid in water in the presence of sufficient ammonia to neutralize 110 to 150% of the carboxylic acid groups in the ethylene-methacrylic acid copolymer for a sufficient
- 10 time to uniformly disperse the ethylene-methacrylic acid copolymer in the water.

12. The process of claim 11 wherein the mixing is carried out at a temperature of about 90 to about 150°C for about 10 minutes to about 2 hours.

**GENERAL POWER OF ATTORNEY**  
(Concerning Several International Patent)

The undersigned, Vernon R. Rice, Vice President and Assistant General Counsel of E. I. DU PONT DE NEMOURS AND COMPANY, 1007 Market Street, Wilmington, Delaware 19898 USA ("DuPont"), hereby confirms that the power to sign for DuPont has been granted to various individuals (as set forth in the attached excerpt from DuPont's Patent Board Rules of Procedure (January 1988), Appendix Section III.A.4), including the Chairman, Vice-Chairman, and those individuals who are Assistant Secretaries of the Patent Board. Currently these Assistant Secretaries are:

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Miriam D. Meconnahey  
Deborah A. Meginniss

In addition, the authority to act on behalf of DuPont before the competent International Authorities in connection with any and all international patent applications filed by it with the United States as Receiving Office and to make or receive payments on its behalf is hereby granted to:

|                        |        |                          |        |
|------------------------|--------|--------------------------|--------|
| Beardell, Lori Y.      | 34,293 | Kuller, Mark D.          | 31,925 |
| Belopolsky, Inna       | 43,319 | Krukiel, Charles E.      | 27,344 |
| Benjamin, Steven C.    | 36,087 | Jarnholm, Arne R.        | 30,396 |
| Birch, Linda D.        | 38,719 | Langworthy, John A.      | 32,255 |
| Bowen, Jr., Alanson G. | 24,027 | Lerman, Bart E.          | 31,897 |
| Christenbury, Lynne M. | 30,971 | Levitt, Cary A.          | 31,848 |
| Cotreau, William J.    | 36,490 | Li, Kening               | 44,872 |
| Deitch, Gerald E.      | 30,457 | Magee, Thomas H.         | 27,355 |
| Deshmukh, Sudhir       | 33,677 | Mayer, Nancy S.          | 29,190 |
| Dobson, Kevin S.       | 40,296 | Medwick, George M.       | 27,456 |
| Duffy, Roseanne R.     | 33,869 | Morrissey, Bruce W.      | 30,663 |
| Edwards, Mark A.       | 39,542 | Reynolds, Stephen E.     | 37,580 |
| Estrin, Barry          | 26,452 | Rizzo, Thomas M.         | 41,272 |
| Evans, Craig H.        | 31,825 | Santopietro, Lois A.     | 36,264 |
| Fair, Tamera L.        | 35,867 | Schaeffer, Andrew L.     | 33,605 |
| Feltham, S. Neil       | 36,506 | Sebree, Chyrrea J.       | 45,348 |
| Floyd, Linda Axiemethy | 33,692 | Shafer, Robert J.        | 24,437 |
| Frank, George A.       | 27,636 | Shay, Lucas K.           | 34,724 |
| Golian, Andrew G.      | 25,293 | Shipley, James E.        | 32,003 |
| Gorman, Thomas W.      | 31,959 | Siegeil, Barbara C.      | 30,684 |
| Gould, David J.        | 25,338 | Sinnott, Jessica M.      | 34,015 |
| Griffiths, John E.     | 32,647 | Steinberg, Michael A.    | 43,160 |
| Hamby, Jane O.         | 32,872 | Steinberg, Thomas W.     | 37,013 |
| Hamby, William H.      | 31,521 | Stevenson, Robert B.     | 26,039 |
| Heiser, David E.       | 31,366 | Strickland, Frederick D. | 39,041 |
| Hendrickson, John S.   | 30,847 | Tessari, Joseph A.       | 32,177 |
| Joung, J. Kenneth      | 41,881 | Tulloch, Rebecca W.      | 36,297 |
| Katz, Elliott A.       | 26,396 | Walker, P. Michael       | 32,602 |
| Kelly, Patricia L.     | 39,247 | Wang, Chen               | 38,650 |
| King, Karen K.         | 34,850 |                          |        |

The undersigned ratifies fully all actions already taken by the above-named individuals in accordance with the authority granted hereby.

E. I. DU PONT DE NEMOURS AND COMPANY

By: \_\_\_\_\_

Vernon R. Rice

Vice President and Assistant General Counsel

Date: \_\_\_\_\_

9-13-00

**DECLARATION and POWER OF ATTORNEY**

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**AQUEOUS DISPERSION COMPOSITION AND MANUFACTURING METHOD FOR THE COMPOSITION**

the specification of which is attached hereto unless the following box is checked:

☒ was filed on **25 FEBRUARY 2000** as U.S. Application No. \_\_\_\_\_ or PCT International Application No. **PCT/US00/04754** and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is known to me to be material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

| Application No. | Country | Filing Date      | Priority Claimed (Yes/No) |
|-----------------|---------|------------------|---------------------------|
| JP48872/1999    | JP      | 25 FEBRUARY 1999 | Yes                       |

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States Provisional Application(s) listed below.

| U.S. Provisional Application No. | U.S. Filing Date |
|----------------------------------|------------------|
|                                  |                  |

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application or PCT International Application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is known to me to be material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

| Application No. | Filing Date | Status (patented, pending or abandoned) |
|-----------------|-------------|---|
|                 |             |   |

**POWER OF ATTORNEY:** I hereby appoint the following attorney(s) and/or agent(s) the power to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

|   |   |
|---|---|
| Name: <b>CRAIG H. EVANS</b>   | Registration No.: <b>31,825</b>   |
| Send correspondence and direct telephone calls to:<br><br><b>CRAIG H. EVANS</b> | <b>E. I. du Pont de Nemours and Company</b><br><b>Legal - Patents</b><br><b>Wilmington, DE 19898, U.S.A.</b><br><br><b>Tel. No.</b><br><b>(302) 992-3219</b><br><br><b>Fax No.</b><br><b>(302) 992-2953</b> |

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

**INVENTOR(S)**

|   |  |                             |                        |
|---|--|-----------------------------|------------------------|
| Full Name of Inventor 1 - <b>NAKATA</b>                   | Last Name                                  | First Name                  | Middle Name            |
|   | <b>NAKATA</b>                              | <b>KAZUYUKI</b>             |                        |
| Signature (please sign full name): <i>Kazuyuki Nakata</i> |  | Date: <i>April 28, 2000</i> |                        |
| Residence & Citizenship                                   | City                                       | State or Foreign Country    | Country of Citizenship |
|   | <b>CHIBA-KEN JPN</b>                       | <b>JAPAN</b>                | <b>JP</b>              |
| Post Office Address                                       | Post Office Address                        | City                        | State or Country       |
|   | <b>2-4-1, YUSHUDAI-NISHI, ICHIHARA-SHI</b> | <b>CHIBA-KEN</b>            | <b>JAPAN</b>           |
|   |  |                             | Zip Code               |

☐ Additional Inventors are being named on separately numbered sheets attached hereto.